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Permeability of Polymeric Membrane Lining Materials

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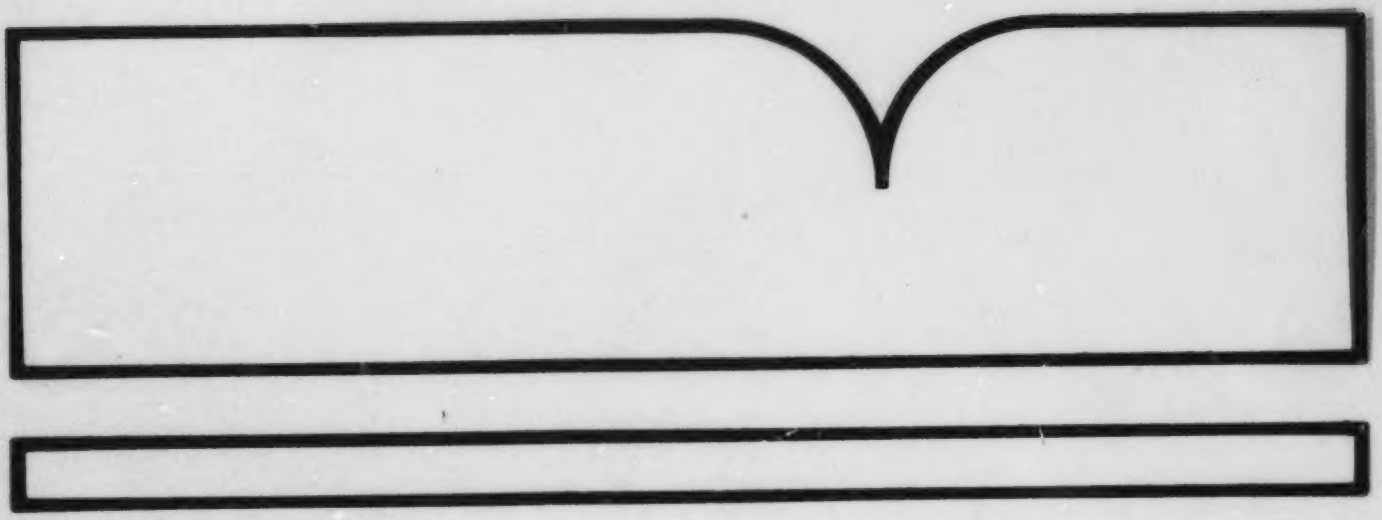

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PERMEABILITY OF POLYMERIC MEMBRANE LINING MATERIALS

by

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16. ABSTRACT

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PERMEABILITY OF POLYMERIC MEMBRANE LINING MATERIALS

Permeabilities to three gases (carbon dioxide, methane, and nitrogen), water vapor, and five solvents (methanol, acetone, cyclohexane, xylene, and chloroform) are reported for a broad range of commercial polymeric membranes. Gas and water vapor transmission (WVT) data were determined by ASTM methods and solvent vapor transmission data were obtained in a modified WVT procedure. Permeability characteristics of thermoplastic and partially crystalline membranes were also assessed in pouch-type tests with salt solutions, actual wastes, acetone, xylene, and water-soluble and organic-soluble dyes. All membranes were permeable to some extent. Magnitude and direction of fluid transport vary with the membrane composition and its thickness, temperature, the permeant fluid, and the driving force which depends upon the concentration or the vapor pressure gradient across the membrane. Permeability depends upon the solubility and diffusion characteristics of the permeant in the particular membrane.

INTRODUCTION

Because of their low permeability to gases, liquids, and vapors, flexible polymeric membranes offer ~~the best overall~~ a feasible approach to meeting the requirements of the U.S. Environmental Protection Agency's regulations for the lining of waste storage, treatment, and disposal facilities to prevent migration into the groundwater of waste liquids and their constituents. In the initial use of liners for waste disposal facilities, the primary purpose was to control the transmission of water and trace metals to the groundwater. To these species polymeric membranes have shown extremely low permeation. However, polymeric compositions are well known to be permeable to organic species (7, 8), and differ greatly in magnitude, depending on the polymeric material and the permeating fluid or molecular species. The migration of organics, which are of great concern in groundwater protection, should also be controlled by membrane liners. From the standpoint of designing lined hazardous waste facilities, it is important to know the magnitude of the permeability of the polymeric membranes to different constitutents and to know the driving forces that are involved so that transmission rates can be estimated.

This paper discusses the permeability of polymeric membrane liners and presents experimental results for a range of commercial polymeric membranes to gases, water vapor, and solvent vapors. Also, presented are current results of studies of the permeability characteristics of membranes as observed in pouch experiments.

TRANSPORT THROUGH POLYMERIC MEMBRANES

In contrast to soils, polymeric membrane liners are nonporous. In soils the flow of liquids containing dissolved constituents is through the pores with hydraulic gradient the principal driving force. For nonporous membranes, the transport of a permeating fluid, be it gas, liquid, or vapor, depends on the solubility of the constituent in the membrane and its diffusibility in the membrane. This transport proceeds essentially in three steps:

1. Dissolution of the fluid or constituent into the membrane.
2. Diffusion of the fluid through the membrane.
3. Evaporation or dissolution of the fluid on the downstream side of the membrane.

Step 1 depends upon the solubility of the fluid in the membrane and relative "activity" of the fluid on both sides of the interface. The diffusion through the membrane in Step 2 involves a variety of parameters which include the molecular size of the permeating fluid and various characteristics of the membrane compound, e.g. presence of fillers, crystalline zones, plasticizers, crosslinks, etc. Step 3 is similar to the first step and depends upon the relative "activity" of individual constituents on both sides of the interface at the liner surface.

The major driving force for the movement of a given constituent through a membrane is its concentration gradient across that membrane. Each constituent in a gaseous or liquid mixture tends to move independently from a higher to a lower concentration.

GAS PERMEABILITY

Permeability to gases, particularly methane, is an important factor in the use of polymeric lining materials for the control of gas migration in a variety of engineering applications. These include such uses as covers and curtain walls to control movement of methane from landfills and as barriers to prevent entrance of methane into buildings and other structures near MSW landfills.

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The permeability of membrane liner materials to three gases of interest in land disposal facilities was determined by ASTM D1434, Procedure V (1). In this procedure a specimen of the liner is clamped into a stainless steel cell so as to form a barrier to gas flow through the cell. All air is flushed from the system with the test gas and then one side of the cell maintained at a positive pressure while the other remains at atmospheric pressure. A capillary mounted on the atmospheric pressure side of the cell is used to monitor the volume of gas slowly diffusing through the liner. The test apparatus is shown in Figure 1.

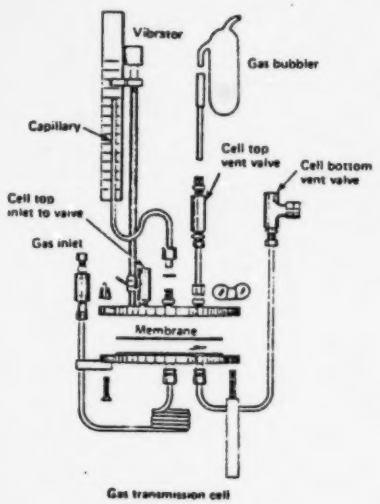

Figure 1. Gas permeability apparatus in ASTM D1434, Procedure V (1).

In determining permeability, both the system pressure and cell temperature must be maintained at constant values throughout the test. Reproducible data cannot be obtained until the test gas, cell, and membrane are at thermal equilibrium and the membrane has reached equilibrium in its absorption of the test gas. When these conditions are met, the passage of test gas through the membrane will reach a steady state rate and the permeability determination is made.

Data collection consists of tabulating the linear displacement of the liquid meniscus within the capillary versus time. This data can be reduced to a displacement rate using a linear regression routine and further reduced to the gas transmission rate (GTR) in terms of gas volume/area x time x Δ pressure if the capillary diameter, specimen surface area, and applied pressure differential are recorded. Determination of the gas transmission rate at two or more temperatures allows the calculation of an activation energy (E_d) and of a preexponential factor (log GTR_0) for the transmission process. Knowledge of these two parameters for a given liner-gas combination makes possible the calculation of the gas transmission rate at any temperature.

Figure 2 illustrates the relation between temperature and transmission rate of carbon dioxide, nitrogen, and methane through an elasticized polyolefin (ELPO) membrane. This relationship is roughly linear over a moderate temperature range.

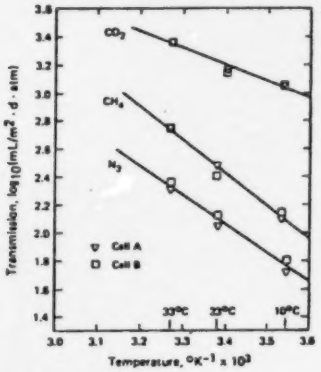

Figure 2. Permeability of ELPO to CO_2, CH_4, and N_2 as a function of temperature. Data are reported as GTR for a membrane of 0.158 mm under pressure difference of one atmosphere.

The data for CO_2 are tabulated in Table 1.

TABLE 1. TEMPERATURE DEPENDENCE OF LINER PERMEABILITY TO CARBON DIOXIDE

Cell	Temperature, °C	$(T°K)^{-1}$ X values	GTR, mL(STP)/m^2·d·atm	$\log_{10}$ GTR Y values
A	10	3.531×10^{-3}	1120	3.0492
A	23.1	3.395×10^{-3}	1140	3.1492
A	33.0	3.266×10^{-3}	2290	3.3598
B	10	3.531×10^{-3}	1150	3.0607
B	23.1	3.395×10^{-3}	1490	3.1732
B	33.0	3.266×10^{-3}	2330	3.3674

Data for the permeability of a series of polymeric membrane liners to carbon dioxide, methane, and nitrogen at 23°C are presented in Table 2.

These data presented on gas permeability show:

1. Major differences in permeability among the lining materials.
2. Permeability of polymeric materials differs for a given generic polymer type due to compound variation, (e.g. filler and plasticizer contents).
3. Rates of transmission varied with the gas.
4. Gas transmission decreased with increased thickness of the membrane.

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5. Higher polymer crystallinity yields lower permeability, as shown when comparing LDPE, LLDPE (linear low-density polyethylene), and HDPE.

6. Permeability of polymeric membranes to gases increases with temperature.

TABLE 2. PERMEABILITY OF POLYMERIC MEMBRANE LINERS TO GASES AT 23°C DETERMINED BY ASTM D1434, PROCEDURE V

Polymer	Liner serial number[a]	Thickness, mm	Type[b]	Gas transmission rate, mL(STP[c])/m²·d·atm CO_2	CH_4	N_2
Butyl	44	1.60	XL	512	120	19.7
CPE	77	0.72	TP	106[d]	6.31[d]	≤1.45[d]
CSPE	6R	0.82	TP-FR	122	21.6	26.2
CSPE	55	0.86	TP	418	124	27.1
ELPO	36	0.58	CX	1450	280	125
EPDM	83R	0.89	TP-FR	2720[e]	...	...
EPDM	91	0.90	XL	5260	1400	314
EPDM	8	1.50	XL	...	470[f]	...
Neoprene	90	0.90	XL	716	80.9	31.1
PB	221	0.71	CX	818	248	62.3
HDPE	265	0.61	CX	729	138	...
HDPE	269	0.86	CX	467	104	...
LDPE	21	0.25	CX	6180[e]	1340[e]	...
LLDPE	281	0.46	CX	1370	322	...
PVC	93	0.25	TP	7730[e]	1150[e]	
PVC	88	0.49	TP	3010	446	108
PVC	59	0.81	TP	2840[e]	285[e]	...

[a]Matrecon liner serial number; R = Fabric-reinforced.
[b]XL = Crosslinked; TP = Thermoplastic; CX = Crystalline; FR = Fabric-reinforced.
[c]STP = Standard temperature and pressure.
[d]Measured at a pressure gradient of 40 psi; all other measured at 20 psi.
[e]Measured at 30°C.
[f]Measured at 20°C.

WATER VAPOR TRANSMISSION (WVT)

The permeability of polymeric membranes to water vapor is important to a variety of applications such as the covers for reservoirs and other impoundments, the lining of canals and tunnels, and as a moisture barrier in various buildings and structures.

The WVT rates of polymeric membrane liners were determined by ASTM E96, Procedure BW (2). In this procedure, a circular specimen of the liner is sealed with molten wax into the mouth of an aluminum cup partially filled with deionized water. The test cell is illustrated in Figure 3. The entire assembly is kept in an inverted position so that water is in contact with the membrane surface and stored in a cabinet maintained at 23°C and a relative humidity (RH) of 50 ±5% and equipped with a small fan to ensure uniform air velocity over the surfaces of the specimens as required by the procedure. The WVT occurs across a water vapor pressure gradient of 100% RH (inside the cup) to 50% RH.

The test cells are periodically weighed and the resulting data are reduced using a linear regression program to yield a loss rate which can be converted to a WVT value in units of g/m²·d. WVT data for a series of polymeric membrane liners are presented in Table 3 by polymer and increasing thickness.

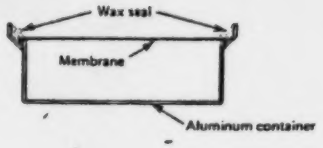

Figure 3. Cross section of water vapor transmission cup used in ASTM E96 (2).

TABLE 3. WATER VAPOR TRANSMISSION (WVT) RATES OF POLYMERIC MEMBRANE LINERS

Polymer	Liner serial number[a]	Thickness, mm	Type	WVT, g/m²·d
Butyl rubber	163R	0.85	XL-FR	0.384
	57	0.85	XL	0.020
	22	1.85	XL	0.097
CPE	86	0.53	TP	0.643
	135	0.79	TP	1.400
	77	0.79	TP	0.320
	12	0.85	TP	0.264
	147R	0.94	TP-FR	0.305
	165	0.97	TP	0.643
CSPE	169R	0.74	TP-FR	0.333
	148	0.76	TP	0.663
	55	0.89	TP	0.438
	151R	0.91	TP-FR	0.748
	6R	0.94	TP-FR	0.422
	170R	1.07	TP-FR	0.252
ELPO	36	0.72	CX	0.142
ECO[b]	178	1.160	XL	20.18
	178	1.650	XL	14.30
EPDM	41	0.51	XL	0.270
	83	0.94	XL	0.190
	8	1.70	XL	0.172
Neoprene	42	0.51	XL	0.304
	168	0.91	XL	0.473
	167R	1.27	XL-FR	0.429
	9	1.59	XL	0.237
Nitrile rubber	171R	0.76	TP-FR	5.51
PB[c]	221	0.69	CX	0.084
PEEL[d]	75	0.20	CX	10.50
LDPE	108	0.76	CX	0.0573
HDPE	184	0.80	CX	0.0172
	179	2.44	CX	0.0062
HDPE-A	181	0.86	CX	0.0472
PVC	89	0.28	TP	4.42
	17	0.51	TP	2.97
	146	0.76	TP	1.94
	143	0.79	TP	1.85
PVC-E	176R	0.91	TP-FR	2.78
PVC-OR	40	0.83	TP	4.17
Saran film (0.5 mil)	222	0.013	TP	0.563

[a]Matrecon serial number; R = fabric-reinforced.
[b]ECO = epichlorohydrin rubber.
[c]PB = polybutylene.
[d]PEEL = polyester elastomer.

As with the gas transmission data, WVT varies considerably among the polymer types; for example, the rates are much lower through hydrocarbon types (e.g. butyl rubber, EPDM, ELPO), than through polar types (e.g. ECO, nitrile rubber). Also, within a polymer type there is considerable variation due

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to differences in composition. Increased thickness and increased crystallinity, in the case of crystalline materials, reduce transmission rates. Under the conditions of this test, a loss of one $g/m^2 \cdot d$ is equal to 1.07 gallon per acre per day.

SOLVENT VAPOR TRANSMISSION (SVT)

Considerable data (7, 8) exist with respect to the transport of organics through polymeric films, but almost no data exist with respect to polymeric membrane liners. Preliminary experiments were performed on neat solvents to assess their transmission rates under controlled conditions through different membranes. Solvent vapor transmission rates were determined by a modification of ASTM E96 in which a circular specimen of the liner is mechanically clamped onto the mouth of an aluminum cup partially filled with the test solvent (Figure 4). The method differs from E96, Procedure BW, in that the cups are stored in an upright position so that solvent vapor contacts the membrane. SVT occurs as a result of the concentration gradient across the membrane by the presence of a saturated atmosphere within the cup and the essentially zero level outside the cup. The rate of SVT is determined in the same manner as described above for WVT.

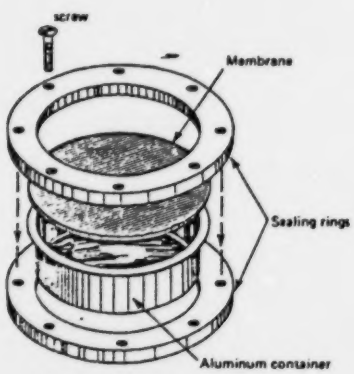

Figure 4. SVT cup with aluminum sealing rings.

SVT data for a series of membranes selected for good solvent resistance are given in Table 4. These data are for five organic solvents, i.e. methanol, acetone, cyclohexane, xylene, and chloroform. Although limited, the data show substantially different transmission rates among the membranes and among the different solvents. Increased crystallinity among the polyethylene membranes reduces transmission, as does increased thickness. The alloying of HDPE increases transmission.

PERMEABILITY OF MEMBRANES IN POUCH TESTS

The permeability characteristics of thermoplastic and partially crystalline polymeric membrane have been studied in a pouch procedure which simulates some in-service conditions of liners in waste disposal facilities (3, 4, 5, 6,). In this procedure, small containers are fabricated of the membrane to be tested and filled with a test liquid, sealed, and immersed in a liquid of known composition. When the outer liquid initially is DI water, permeabilities of the membrane to water and to the dissolved constituents can be determined by observing with time the change in the weight of the pouch and changes in the pH, electrical conductivity, and the composition of the outer water. Because of differences in the concentrations of different molecular species across the membrane wall, osmosis will cause the outer water to enter the pouch and cause ions and dissolved constituents in the pouch liquid to tend to leave the pouch and enter the outer water. An increase in weight of the pouch may only reflect absorption of the outer water by the wall and not permeation of water into the pouch. When the test is completed the pouch and its contents are weighed separately to determine the actual amounts that have been absorbed by the wall and the amounts that have permeated the pouch wall.

TABLE 4. SOLVENT VAPOR TRANSMISSION (SVT) RATES OF POLYMERIC MEMBRANE LINERS BY ASTM E96, MODIFIED

Polymer	Liner serial number	Thickness, mm	Type	SVT, $g/m^2 \cdot d$				
				Methyl alcohol	Acetone	Cyclohexane	Xylene	Chloroform
CSPE	170R	1.07-1.12	TP-FR	...	221	...	...	...
ELPO	172	0.53-0.61	CX	2.10	8.62	760	359	3230
HDPE	184	0.77-0.83	CX	0.16	0.56	11.7	21.6	54.8
	179	2.42-2.81	CX	...	...	...	6.66	15.8
HDPE-A	180	0.53	CX	...	...	...	295	...
	181	0.85-0.88	CX	0.50	2.19	151	124	506
	182	0.97	CX	...	...	...	220	...
LDPE	108	0.74-0.76	CX	0.74	2.83	161	116	570
PB	221	0.64-0.74	CX	0.35	1.23	616	178	2120
Teflon	234	0.10	FEP-Y	0.34	1.27	0.026	0.16	20.6

Experimental Results

The studies of the permeability of polymeric membranes using the pouch procedure included membranes of CPE, CSPE, HDPE, HDPE-A, LDPE, PB, PVC, and PVC-OR (4). The test liquids included aqueous salt solutions (sodium chloride and lithium chloride), organic solvents (acetone and xylene), mixtures of acetone and water, water-soluble and organic-soluble dyes, and several actual waste liquids.

Permeability to Water

Of nine pouches filled with the 5% NaCl aqueous solution, eight showed the transmission of water into the pouch, as judged by the increased weight of the pouch contents (Table 5). The ninth pouch, CSPE 55, gained in weight but showed a weight loss for its contents indicating absorption of the pouch liquid by the wall. The pouches with a highly acidic waste showed the transmission of water from outside to inside the pouches. The weight changes during exposure are shown in Figure 5. Table 6 presents the weight data on the pouches when they were dismantled. The results indicate a low permeability for ELPO. The behavior of pouches containing a highly alkaline waste was similar except for the ELPO pouch which gained weight after 300 days of exposure, indicating a slowly developing effect of the waste on this membrane.

The effect of the concentration gradient across the pouch wall is clearly demonstrated in the PVC pouches with LiCl (Figure 6). The pouches containing 5% LiCl show a rate of transmission that is approximately half that of the pouches with 10% LiCl.

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TABLE 5. POUCH TEST OF MEMBRANES WITH 5% NaCl AS THE TEST LIQUID

Polymer[a]	Exposure time, d	Weight change, g: Empty pouch	Filled pouch	Pouch contents	Water transmission, g/m2d	EC outer water[b], µmho
CPE 86	316	3.60	4.02	0.42	0.291	86
CSPE 6R	1151	6.97	10.23	3.26	0.069	585
CSPE 55	315	5.35	4.70	-0.65	-0.002	66
ELPO 36	1151	-0.01	2.32	2.31	0.049	34
LDPE 21	315	-0.04	0.85	0.81	0.073	31
PB 98	316	0.14	0.65	0.51	0.043	27
PVC 17	310	-0.40	6.45	6.85	0.619	371
PVC 19	315	-0.60	1.77	2.37	0.214	16
PVC 59	1115	0.19	11.00	10.81	0.238	17

[a]Number = Matrecon serial number; R = fabric-reinforced.
[b]Electrical conductivity of outer water before it was replaced at 1169 days.

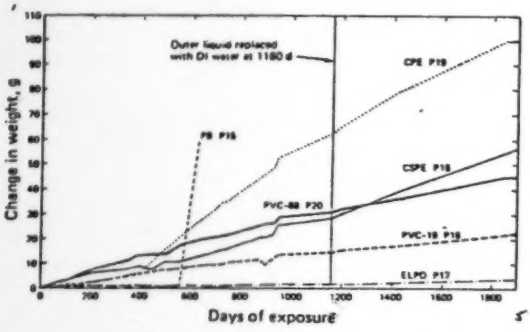

Figure 5. Weight changes of pouches containing a highly acidic waste.

TABLE 6. POUCH TEST OF MEMBRANES WITH A HIGHLY ACIDIC WASTE AS TEST LIQUID

Polymer[a]	Exposure time, d	Weight change, g: Empty pouch	Filled pouch	Pouch contents	Water transmission, g/m2d	EC outer water[b], µmho
CPE 86	1887	14.28	100.00	98.70	1.213	14,600
CSPE 85	1887	28.26	58.87	46.57	0.510	2,500
ELPO 36	1887	1.03	3.58	1.40	0.017	92
PVC 19	1887	1.94	22.50	21.03	0.252	4,800
PVC 88	1887	11.46	45.43	35.84	0.363	9,000

[a]Number = Matrecon serial number; R = fabric-reinforced.
[b]Electrical conductivity of outer water before it was replaced at 1169 days.

The pouches filled with acetone or xylene and placed in DI water showed the movement of each solvent out of the pouches and no water movement into the pouches (Figures 7 and 8). The pouches with 50:50 acetone:DI water showed outward movement of acetone when immersed in DI water and inward movement of acetone when immersed in acetone (Figure 9).

Permeability to Ions

In the experiments performed with salt solutions, the ions showed little tendency to permeate polymeric sheetings. Even at high concentration gradients (10% LiCl solution), the electrical conductivity of the outer liquids remained low. In the tests with the highly acidic waste, the hydrogen ions in the pouches

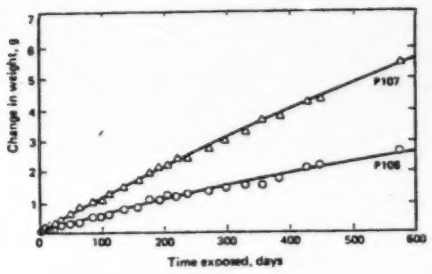

Figure 6. Weight changes of PVC pouches containing 5 (P106) and 10% (P107) aqueous solutions of LiCl during immersion in DI water.

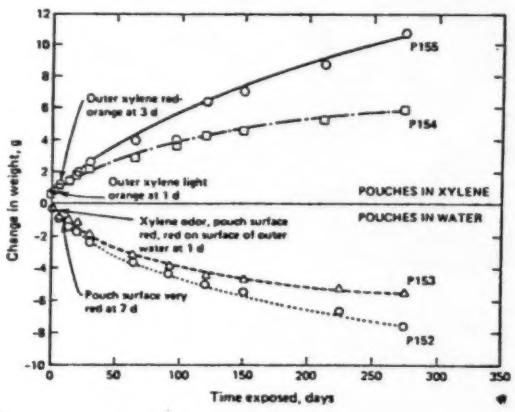
P155
P154
Outer xylene red-orange at 3 d
Outer xylene light orange at 1 d
POUCHES IN XYLENE
POUCHES IN WATER
Xylene odor, pouch surface red, red on surface of outer water at 1 d
P153
Pouch surface very red at 7 d
P152
Change in weight, g
Time exposed, days

Figure 7. Weight changes in HDPE-A pouches filled with xylene and 1% Automate Red.

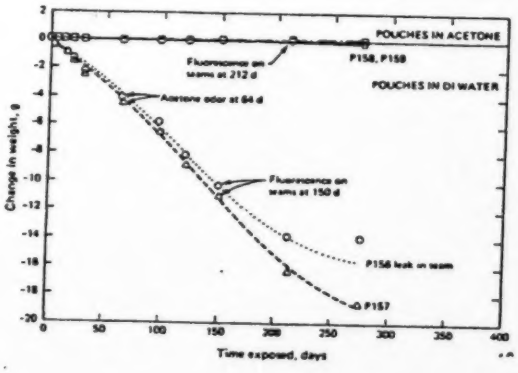

Figure 8. Weight changes in HDPE-A pouches filled with acetone and 1% Fluorescent Yellow.

appeared to have migrated through the walls; the pH of the outer solution dropped and the electrical conductivity increased (Table 6).

PERMEABILITY OF POLYMERIC MEMBRANE LINING MATERIALS

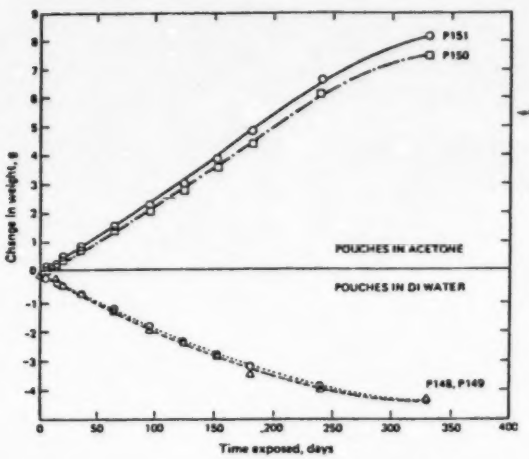

Figure 9. Weight changes in HDPE-A pouches with 50:50 acetone:DI water and 1% methyl violet.

Permeability to Organics

Both acetone and xylene permeated the walls of the pouches (Figures 8, 9, and 10) when the pouches were placed in DI water. The acetone permeated the walls and dissolved in the outer water; the xylene permeated but, because it is not soluble in water, rose to the surface. When the pouches with the acetone and xylene were placed in the same solvents, the movement was into the pouch where the solvent contained dissolved constituents, either organic dyes or water (Figures 8-10). Pouches with waste liquids that were oily developed a film of oil on their outside surfaces.

Permeability to Organic Dyes

Three water-soluble and two organic-soluble dyes were studied in pouch experiments at 1% concentrations. The water-soluble types included sodium fluorescein, Sevron Red, and methyl violet. The sodium fluorescein showed positive transmission through PVC-OR, and trace transmission through HDPE, PB, and PVC. Sevron Red in water showed no detectable transmission through HDPE-A and PB. Methyl violet in the 50:50 acetone:water mixture showed no detectable transmission through HDPE-A. In contrast to the water-soluble dyes, the two organic-soluble dyes, Automate Red and Fluorescent Yellow, in pouches containing acetone and xylene quickly permeated the HDPE-A pouch walls (Figure 7). For pouches immersed in DI water, these organic dyes precipitated on the outside as they are insoluble in water.

Comments on the Pouch Procedure

A major feature of the pouch procedure is its ability to assess, over extended periods, the permeability of a liner material simultaneously to several waste constituents in a configuration that simulates some aspects of the membrane liner in service in a disposal facility.

PERMEATION OF ORGANICS THROUGH MEMBRANES IN SOIL ENVIRONMENTS

In actual service a membrane liner is placed on a porous layer (e.g. soil or geotextile), the permeability of which can range greatly. Also, a liner often is covered or becomes covered with soil, sand, silt, or sludge. These materials will tend to retard the transmission of waste constituents, as has been demonstrated in preliminary experiments.

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